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Monitoring Spent or Reprocessed Nuclear Fuel Using Fast Neutrons

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Over the past few years a number of experiments have been carried out at LLNL with a scintillator array that has the ability to count individual MeV neutrons and γ-rays with nanosecond timing. It has been demonstrated that this array can be used to measure the statistical properties of the neutrons emitted in single fission chains. The multiple time scales over which these fission neutrons are correlated allow one to deduce quite a lot regarding the nature of the fissile assembly. In this paper we will describe how neutron correlations measured with a liquid scintillator array can be used to assay the amounts of fissile elements in reprocessed and spent nuclear fuels.

I. Historical Background

The basic theory of neutron fluctuations and time correlations in near critical assemblies of fissionable materials was developed during WWII by R. Feynman and E. Schrödinger. The most important prediction is that a modest amount of multiplication leads to long fission chains and highly non-Poisson neutron count distributions. Feynman and de Hoffmann (1944). developed a formula for relating the excess neutron intensity fluctuation relative to Poisson distribution

$$\frac{<c^2 - \bar{c}^2>}{\bar{c}} - 1 \equiv 2Y_{2F}$$

in a homogeneous sub-critical assembly to the neutron multiplication M and neutron lifetime $1/\alpha$ inside the assembly. In particular, they showed that

$$Y_{2F} = \varepsilon\left(M - \frac{M-1}{\bar{\nu}}\right)\left(\frac{\nu_{2s}}{\bar{\nu}_s} + (M-1)\frac{\nu_{2i}}{\bar{\nu}_i}\right)$$
$$* \left[1 - \frac{1 - e^{-\alpha t}}{\alpha t}\right], \quad (1)$$

where ε is the detector efficiency, and ν and ν_2 are the average numbers of neutrons and neutron pairs in a single fission[1]. The indices *s* and *i* refer to spontaneous and induced fissions. This formula agreed with measurements of Y_{2F} for the "Water Boiler Reactor" carried out at Los Alamos in 1944 by E. Segre.

If the detector efficiency ε is unknown, then Eq. (1) must be supplemented with additional information in order to determine the multiplication. One possibility is to use the 3-neutron Feynman correlation function Y_{3F} (Ref 2). In the decades following WW II determining the multiplication of sub-critical assemblies using ^{3}He neutron detectors to measure the 2- and 3-neutron correlation functions Y_{2F} and Y_{3F} became a cottage industry at LANL. However, because ^{3}He neutron detectors can only see thermal neutrons it is difficult to distinguish individual fission chains or distinguish the effects of multiple time scales in a non-homogeneous assembly. These deficiencies have been overcome with development of neutron detectors that can directly detect fission neutrons with nanosecond timing.

II. Fast Neutron Counting

In 2008, W. Stöffl et al built a liquid scintillator array that uses liquid xylene and has the ability to count individual MeV neutrons and γ-rays with nanosecond timing resolution[3]. A number of experiments have been carried out with this array and its descendants that demonstrate its usefulness for directly seeing the multiple neutrons emitted in a single fission chain.
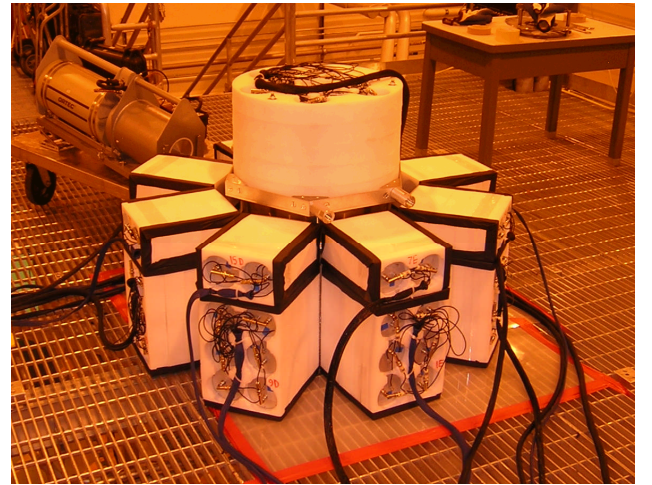

Fig. 1. Liquid scintillator array with fast electronic readout used in LLNL experiments

One type of problem where fast neutron counting has turned out to be particularly useful is the analysis of non-homogeneous fissile material assemblies, for example a fissile material assembly surrounded by a layer of moderating material. The neutron multiplication M of a fissile assembly surrounded by an external moderator will be larger than that of the "bare" fissile assembly because neutrons that are thermalized in the external moderator can be reflected back into the fissile assembly, producing additional fission chains. If the assembly contains isotopes with a large thermal neutron cross-section, e.g. ^{235}U or ^{239}Pu, the increase in the multiplication can be significant. The Feynman 2-neutron correlation function including these "restart" fissions is given by:

$$Y_{2F} = \left(\frac{M_0}{M}\right)^2 Y_{2F}(\alpha_1 t) + \left(1 - \frac{M_0^2}{M^2}\right) Y_{2F}(\alpha_2 t), \qquad (2)$$

where the $Y_{2F}(\alpha t)$ functions are similar in form to the homogeneous Feynman-de Hoffmann formula (1). This formula works particularly well in cases where the time scales $1/\alpha_1$ and $1/\alpha_2$ are very different. This happens to be the case, for example, when a homogeneous fissile material is surrounded by a distinct thin moderating shell.

III. Assaying the Pu in MOX Fuel

One example of a situation where one might hope that the formula (2) is useful is a nuclear fuel assembly surrounded by a shell of moderating material such as polyethylene. We have obtained evidence that this expectation is correct by carrying out Monte Carlo calculations using MCNP for simulated fuel assemblies surrounded by a cylindrical array of liquid scintillators.
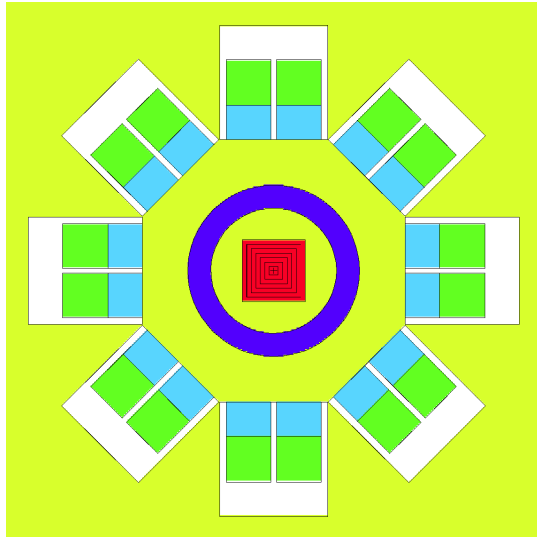

Fig. 2. Geometry used in Monte Carlo calculations.

The geometry used in the calculations is shown in Fig. 2. We assumed that the nuclear fuel (indicated in red)

has a square cross-section of side 14 cm and is 1 m long. The polyethylene moderator (indicated in blue) was assumed to be a cylindrical shell 5 cm thick over the length of the assembly. The cylindrical detector array was 1.4 m long and contained 192 10-cm diameter, 7.5-cm deep xylene cells. The actinide compositions used in our simulations are shown in Table 1. The observing time was typically 100 seconds.

We obtained the 2-neutron correlation function Y_{2F} by analyzing the statistics of the fast neutrons recorded in the cylindrical stack of liquid scintillators. Our strategy is to use the simulated 2-neutron and 3-neutron correlations for time intervals less than a microsecond to determine the rate of generation of spontaneous neutrons and the "bare" multiplication M.

In Fig 3 we show the results of a Monte Carlo calculation of the Feynman correlation function Y_{2F} for 125kg of MOX fuel with no CH_2 layer.
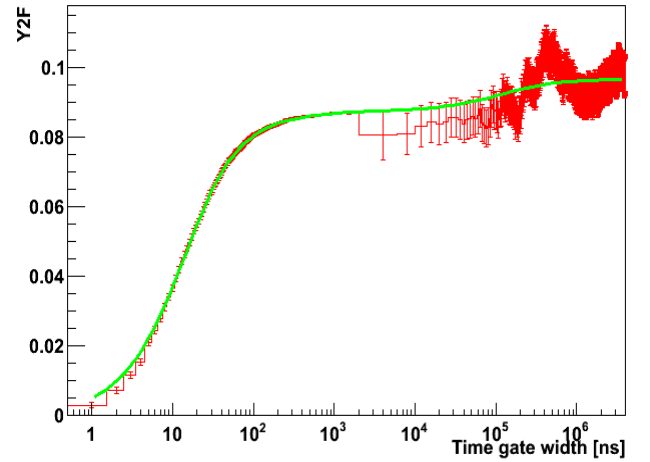

Fig. 3. Monte Carlo Y_{2F} for 125 kg MOX fuel with no surrounding CH_2 moderator.

Our simulated MOX fuel contained no ^{242}Cm or ^{244}Cm, so the predominant source of spontaneous fission neutrons is ^{240}Pu. The values for the detector efficiency and "bare" multiplication we obtained from our Monte Carlo calculations of Y_{2F} (shown in Fig. 3) and Y_{3F} for the 125 kg of MOX fuel with no external CH_2 moderator are $\varepsilon = 7.5\%$ and $M_o = 1.17$. The source strength of spontaneous fission neutrons we infer from the observed count rate, together with the inferred values of M_o and ε, is completely consistent with the spontaneous fission rate expected from the 0.6 kg of ^{240}Pu in 125 kg of MOX fuel.

Given M_o the amount of $^{235}U + ^{239}Pu + ^{241}Pu$ in the MOX fuel can be inferred from the behavior of the 2-neutron correlation function Y_{2F} for time intervals on the order of milliseconds when the fuel assembly is surrounded by an external CH_2 moderator. Fig 4 shows the 2-neutron correlations we obtained from our Monte

Carlo calculations when the MOX fuel is surrounded by a 5-cm thick CH_2 layer. Assuming $M_o = 1.17$, the detector efficiency and total multiplication M obtained from the Monte Carlo calculation of Y_{2F} for the MOX fuel with a 5cm CH_2 layer are $\varepsilon = 3.5\%$ and $M = 1.54$. It is expected the bare multiplications are the same in the no moderating layer and moderated cases. Again, the source strength inferred from the observed count rate is consistent with the spontaneous fission rate expected from the 0.6 kg of [240]Pu in 125 kg of MOX fuel.
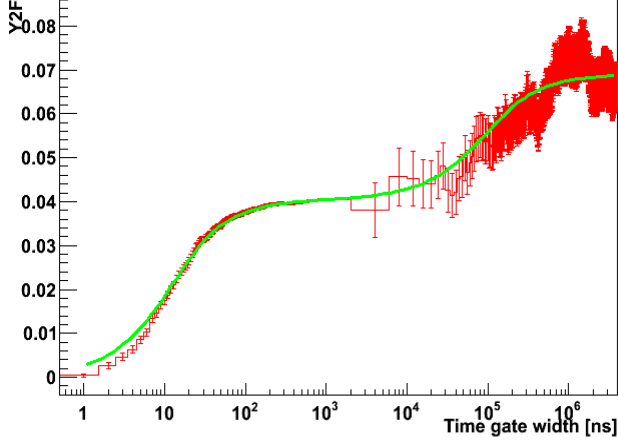
Fig. 4 Monte Carlo Y_{2F} for 125 kg MOX fuel surrounded by a 5 cm CH_2 layer.

The amount of [239]Pu+[241]Pu in the MOX nuclear fuel can be inferred by comparing the asymptotic value for Y_{2F} for moderation times (milliseconds) with the plateau value for Y_{2F} for times on the order of a microsecond. As is evident from Eq. (2), the ratio of these plateau values is $(M/M_0)^2$. The excess of M over M_o is due to the contribution of fissions in the [239]Pu, [241]Pu, and [235]U induced by thermalized neutrons. The value $M = 1.54$ we obtained is consistent with the amounts of these isotopes assumed in the simulation.

From a safeguards perspective, one way of masking the diversion of Pu would be to replace the Pu with a small amount of [252]Cf. If the 4 kg of Pu in our 125 kg of MOX fuel is replaced by 25 µg of [252]Cf, the spontaneous neutron rate will be approximately the same. Figs. 5 and 6 show the results of Monte Carlo calculations for the 2-neutron Feynman correlation functions with 25 µg of [252]Cf replacing the Pu and with and without a 5 cm layer of CH_2. It can be seen that when there is no CH_2 moderator, the Y_{2F} curve is flatter for millisecond times. If we assume that the "bare" multiplication remains the same when the Pu is replaced by Cf, then the multiplication inferred from the ratio of the time > msec plateau to the short time plateau is 1.3 versus 1.54 in the case when Pu is present. The removal of the Pu is reflected in the decrease of the neutron multiplication.
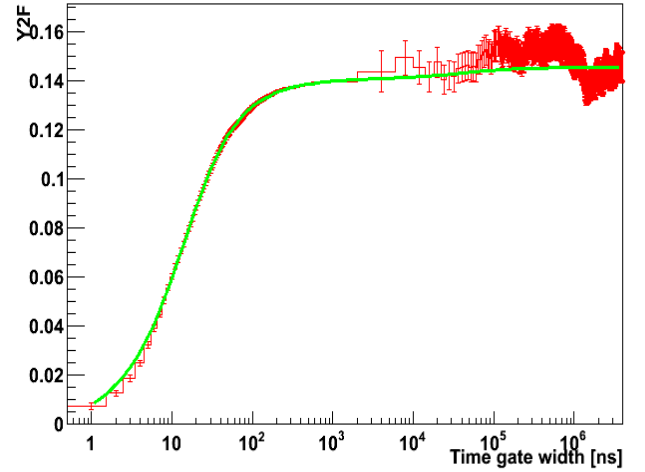

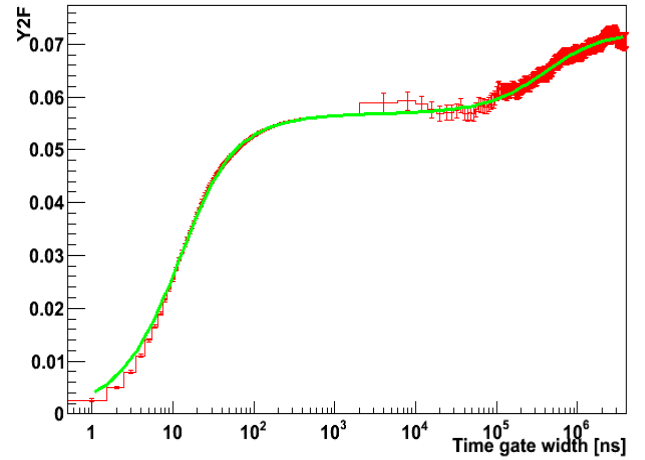Fig. 5 Monte Carlo Y_{2F} for 125 kg MOX fuel with [252]Cf replacing the Pu and no CH_2 layer.

Fig. 6. Monte Carlo Y_{2F} for 125kg MOX fuel with [252]Cf replacing the Pu, but surrounded by a 5 cm CH_2 layer.

TABLE I. Actinide masses used in simulations (kg)

Isotopes	MOX fuel	Spent fuel
[235]U	0.7	20
[238]U	105	125
[239]Pu	3.4	1.4
[240]Pu	0.6	0.5
[241]Pu	0.2	0.2
[241]Am	none	0.04
[242]Cm	none	0.002
[244]Cm	none	0.004

IV. Assaying the Pu and ^{235}U in Spent Reactor Fuel

The case of spent fuel differs from MOX fuel in that the predominant source of spontaneous fission neutrons is ^{242}Cm and ^{244}Cm rather than ^{240}Pu. In addition, a thick high Z shield must be placed between the liquid scintillator array and the fuel in order to shield the liquid scintillator array from the enormous γ-ray background due to the fission products in the spent fuel. Our hope for using neutron correlation techniques to analyze the ^{235}U and Pu content in spent fuel lies in the idea that a high Z shield that is thick enough to reduce the γ-ray flux to acceptable levels, might still be thin enough to measure fission neutron correlations.

In Figs. 7-8, we show the results of Monte Carlo calculations of Y_{2F} and Y_{3F} for a section of a simulated assembly of fuel rods not surrounded by any CH$_2$ with an assumed actinide composition typical for spent reactor fuel with 33000 MWd/ton burn-up,. We assumed that the liquid scintillators could only view a 1 m section of the fuel rod assembly, and that they were shielded with 10 cm of Pb. This thickness of Pb corresponds to about 1 neutron mean free path for fission neutrons.
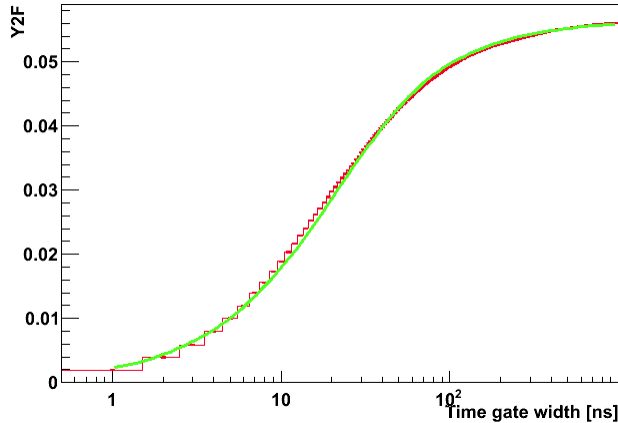

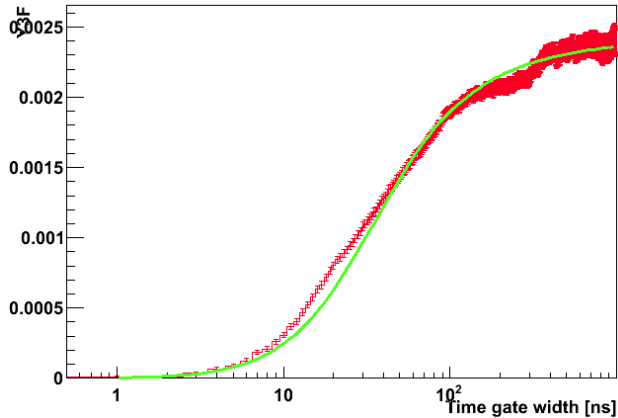Fig. 7. Monte Carlo Y_{2F} for spent fuel shielded with 10 cm Pb and no CH$_2$ layer.

Fig. 8. Monte Carlo Y_{3F} for spent fuel shielded with 10 cm Pb, but no CH$_2$ layer.

As was the case with MOX fuel, we can use Y_{2F} and Y_{3F} to obtain the spontaneous neutron rate and the bare multiplication M_0 of the spent fuel assembly. The detector efficiency ε was determined to be approximately 2% and the bare multiplication M_0=1.1. The observed count rate in this case was 3.5.10^6 sec^{-1}, which is consistent with the assumed amounts of ^{242}Cm and ^{244}Cm.

In Fig. 9, we show the results of Monte Carlo calculations of Y_{2F} when the spent fuel is shielded with 5 cm of CH$_2$ and 10 cm of Pb. The total multiplication M is 1.6. As was the case with MOX fuel, the excess of M over M_0 is due to the fissions induced in ^{239}Pu + ^{241}Pu + ^{235}U by thermal neutrons.

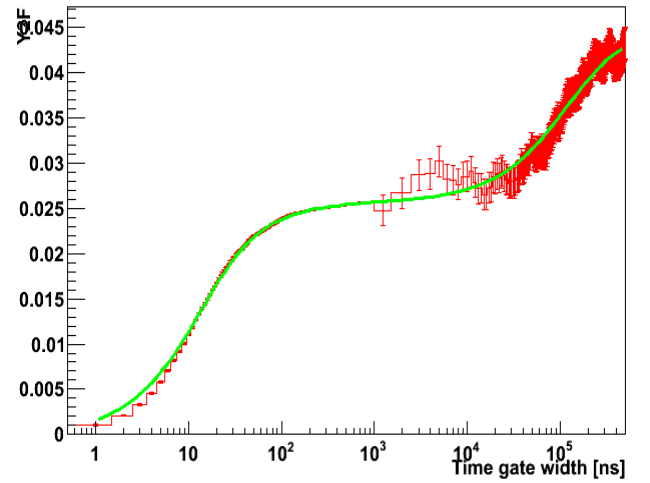Fig. 9. Monte Carlo Y_{2F} for spent fuel shielded with 5 cm CH$_2$ and 10 cm Pb.
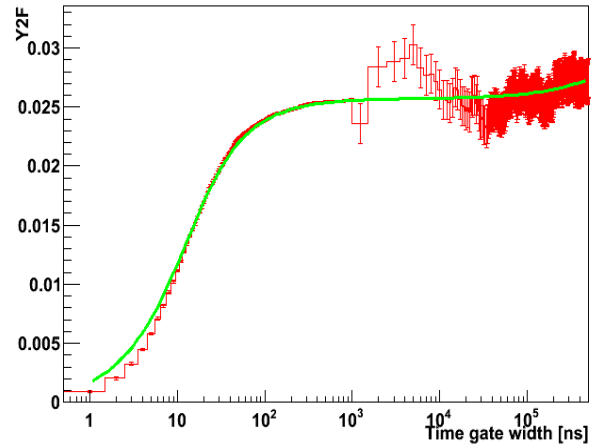

Fig. 10. Monte Carlo Y_{2F} for same fuel configuration, but with the Pu and ^{235}U removed.

In order to get some idea whether it is possible to use our methods to determine the amounts of Pu and ^{235}U remaining in spent fuel rods after they have been removed

from service, we have repeated the spent fuel Monte Carlo calculation with the Pu and ^{235}U removed. The results for Y_{2F} are shown in Fig 10. The multiplication in this case is 1.3 versus 1.6 with no diversion of Pu and ^{235}U. The difference between the average count rate with and without Pu and ^{235}U is a factor 1.5. The ratio of multiplications tells us that $\approx$ 30% of the increase in count rate is due to induced fissions in the ^{239}Pu + ^{241}Pu + ^{235}U. In principle, by using Monte Carlo calculations to map the multiplication M to the combined mass of these isotopes, one could determine this mass by measuring Y_{2F}.

In Fig. 11, we show a calculation of Y_{2F} for the shielded spent fuel with only the Pu removed. The second shoulder in Y_{2F}, is relatively lower than the one in Fig. 9, which indicates that the total multiplication M is reduced. We estimate the total multiplication M in this case to be 1.5 versus 1.6 with no diversion of Pu. Although the results do appear to be sensitive to amount of Pu, it is not clear at this point whether we can determine the individual masses of the ^{239}Pu, ^{241}Pu, or ^{235}U isotopes. This will require further study.
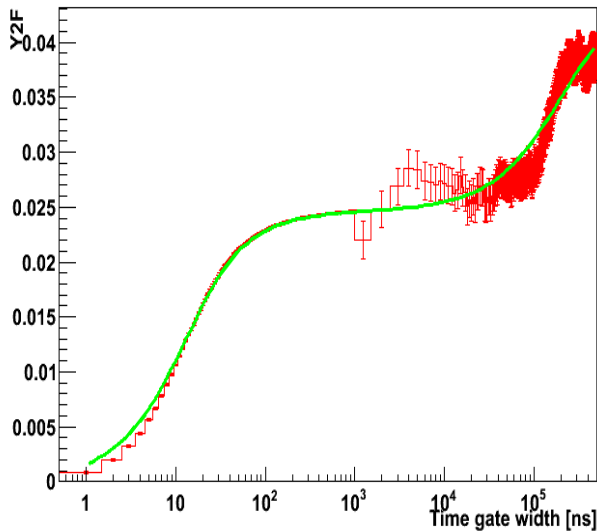

Fig. 11. Monte Carlo Y_{2F} for spent fuel shielded with 5 cm CH$_2$ and 10 cm Pb, but containig no Pu.

V. CONCLUSIONS

Our Monte Carlo simulation results suggest that fast neutron counting techniques ought to be very useful for assaying the amounts of fissile materials in spent and reprocessed nuclear fuel. In particular the short time behavior of Y_{2F} and Y_{3F} can be used to assay the amounts of ^{240}Pu in the reprocessed fuel and Cm in spent fuel. If the reprocessed fuel is surrounded with a few inches of moderator, then the long time behavior of Y_{2F} can be used to quite accurately determine the absolute amount of ^{239}Pu + ^{241}Pu in the reprocessed fuel. In the case of spent fuel

the neutron counting statistics are somewhat marginal due to the need to use a high Z shield thick enough to block the enormous γ-ray flux. However, with a Pb shield about 1 neutron mean free thick, the statistics are good enough to at least estimate the total amount of ^{239}Pu+ ^{241}Pu + ^{235}U in the spent fuel. We are still looking into whether we can identify the amounts of Pu isotopes.

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